RESONANCES IN THE CAPTURE OF PROTONS BY NATURAL SILICON

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MASTER OF SCIENCE
IN
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ABSTRACT

The resonances in the capture of protons by natural silicon were investigated over the range of proton energies from 0.19 to 2.0 Mev. A study of the range from 1.57 to 1.8 Mev revealed prominent resonances at 1.616 and 1.648 Mev with lesser resonances at 1.702, 1.725, 1.774, and 1.818 Mev. Possible resonances were observed in this range at proton energies of 1.582, 1.596, 1.608, 1.633, 1.658, 1.747, and 1.795 Mev. The particular isotope of silicon causing each resonance was not established.

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1. Introduction

The purpose of this project was to investigate and establish the proton energies at which resonance capture occurs when natural silicon is bombarded with protons. The protons were obtained from the Van de Graaff Generator at the U. S. Naval Postgraduate School. Protons of discrete energies in the range of 0.19 to 2.0 Mev were employed.

Natural silicon in the form of SiO was vacuum plated onto a tantalum sheet fastened to a copper target blank. Natural silicon is composed of the following percentages of silicon isotopes.[1]

Isotope	Percentage
Si^{28}	92.21
Si^{29}	4.70
Si^{30}	3.09

The gamma rays resulting from the following reactions were counted by a scintillation counter to establish the resonances:

$$Si^{28}(p,\gamma)P^{29}$$

$$Si^{29}(p,\gamma)P^{30}$$

$$Si^{30}(p,\gamma)P^{31}$$

Simultaneous readings were made with a neutron scintillation counter. While the neutron threshold energies for

[1]General Electric Chart of the Nuclides - Knolls Atomic Power Lab, Fourth Edition

the silicon (p,n) reactions are greater than the proton energies available (Table II), these readings served as a check for neutron-induced reactions which might occur from a contaminated target. No such reactions were found.

2. Historical Background

The establishment of the excited states of nuclei has been the subject of experimental and theoretical investigations since the discovery of natural radioactivity. Through the use of those heavy elements that are naturally radioactive, much information has been obtained.

It was not possible to obtain similar information about light nuclei because none of them have half-lives long enough to be found with natural radioactivity. The advent of various particle accelerators, developed since 1930, has made possible the production of nuclei in various excited states which can be studied. This production has been accomplished by bombarding the nuclei with particles such as protons, alpha particles, electrons, and neutrons. Of these methods of producing radioactive nuclei, only the use of the proton will be discussed since that was the method used in this project.

When a nucleus is bombarded by a proton, one of the events which can occur is resonance capture. Fowler, Lauritsen, and Lauritsen[2] have given an extended analysis of the resonance phenomena. Essentially, the resonance phenomena

[2]W. A. Fowler, et al, Rev. Mod. Phys., 20, 236, (1948)

is as follows: At certain proton energies, the probability is high that the excited compound nucleus, formed by proton bombardment, will be close to one of its quantum excited states. Thus, the probability is high that a compound nucleus will form. The formation of the excited compound nucleus at discrete proton energies is called resonance. The study of resonances has proved valuable in the establishment of the energy levels of excited states of light nuclei. As part of this study, investigations of the capture resonances of the three stable isotopes of natural silicon, Si^{28}, Si^{29}, and Si^{30}, have been conducted.

3. Previous Investigations of Silicon Resonances

The first reports of resonances obtained by bombarding natural silicon with protons were made by Hole, Holtsmark, and Tangen[3]. They reported sharp resonances in the gamma-ray yield for proton energies of 413 and 497 kev.

Later, Tangen[4] conducted further experiments with both thick and thin silicon targets. He obtained the following resonances and attributed the resonances to the indicated isotope:

Resonance	Isotope
326	Si^{29}
367	Si^{30}
414	Si^{29}
499	Si^{30}

[3]N. Hole, et al, Z. F. Phys. 118, 48 (1941).

[4]R. Tangen, Kgl. Norske Videnskab. Selskabs, Skrifter, NR 1 (1946)

The maximum proton energies available to him were 550 kev.

The next reported investigations were made by Seiler[5] in 1955, using the Ohio State University Van de Graaff generator which provided protons with energies up to 1200 kev. The results of his work with natural silicon provided verification of the 414 and 500 kev resonances reported by Tangen. In addition, Seiler reported the following resonances: 622, 675, 698, 703, 732, 760, 778, 944, and 989 kev.

In 1956, Milani, Cooper and Harris[6,7], at Ohio State University, investigated resonances from the Si^{29} $(p,\gamma)P^{30}$ reaction using Si^{29} enriched to 80.8 percent. For this reaction, they report resonance peaks at 326, 414, 698, 731, 918, and 957 kev.

Table I summarizes the reported proton energies at which resonances have been found in the isotopes of silicon. For those resonances where the isotope responsible is not given, positive assignment to Si^{28} or Si^{30} has not been reported to date.

4. Proton Induced Reactions in Silicon

When silicon isotopes are bombarded with protons, there are various nuclear reactions which can occur. One is inelastic scattering in which the silicon nucleus is raised to

[5]M. R. Seiler, M. S. Thesis, Ohio State University, 1955, & Phys. Rev. 99, 340 (1955)

[6]J. N. Cooper, Annual Report by the Ohio State University Research Foundation, March 16, 1955-March 15, 1956. RF Project 440, Report No. 6

[7]S. Milani, et al, Phys. Rev. 99, 645 (1955)

Proton Energy (kev)	Isotope	Bibliography Reference
326	Si^{29}	Ta-1
367	Si^{30}	Ta-1
414	Si^{29}	Ta-1 Se-1 Co-1
499-500	Si^{30}	Ta-1 Se-1
622	Si^{28}	Se-1 Co-1
675		Se-1
698	Si^{29}	Se-1 Co-1
703		Se-1
731-732	Si^{29}	Co-1 Se-1
760		Se-1
778		Se-1
918	Si^{29}	Co-1
944		Se-1
957	Si^{29}	Co-1
989	Si^{28}	Se-1 Co-1

Table I

one of its excited states, followed by the scattering of the proton with reduced energy and the decay of the silicon nucleus to its stable state with the emission of gamma photon(s). Comparison of reported data on the energy levels of silicon[8,9], with resonances obtained during the present investigation, indicates that none of these resonances are attributable to the inelastic scattering process. Other reactions which can occur involve the formation of compound nuclei. Table II lists these reactions and the "Q" value of each reaction. The Q of a reaction is energy difference between the reactants and products of a particular reaction. In mass units, it is given by:

$$Q = (m_t + m_p - m_r - m_e)$$

where m_t is mass of target nucleus

m_p is mass of bombarding particle

m_r is mass of resulting nucleus

m_e is mass of any emitted particle

When the Q value is positive, the kinetic energy of the products is greater than the kinetic energy of the reactants.

From Table II, it is evident that the only reactions which are energetically possible for protons from a 2.0 Mev generator, i.e. reactions with $Q > -2.0$ Mev, are the three (p,γ) reactions.

Based on the above information, the present investi-

[8]D. E. Alburger and E. M. Hafner, Rev. Mod. Phys. 22, 373 (1950)

[9]A. Rubin, et al, Phys. Rev. 100, 961 (1955)

Silicon-Proton Reactions

Reaction	Q(Mev)	Bibliography Reference
$Si^{28}(p,\gamma)P^{29}$	2.724	En-1
$Si^{29}(p,\gamma)P^{30}$	5.5	En-1
$Si^{30}(p,\gamma)P^{31}$	7.290	En-1
$Si^{28}(p,n)P^{28}$	-14.9	Va-1
$Si^{29}(p,n)P^{29}$	-5.749	En-1
$Si^{30}(p,n)P^{30}$	-5.1	En-1
$Si^{28}(p,d)Si^{27}$	-14.976	Se-1
$Si^{29}(p,d)Si^{28}$	-6.246	Va-1
$Si^{30}(p,d)Si^{29}$	-8.388	Va-1
$Si^{28}(p,\alpha)Al^{25}$	-7.70	En-1
$Si^{29}(p,\alpha)Al^{26}$	-4.6	En-1
$Si^{30}(p,\alpha)Al^{27}$	-2.389	En-1

Table II

gation into proton resonance energies was based on the three reactions:

$$Si^{28}(p,\gamma)P^{29}$$
$$Si^{29}(p,\gamma)P^{30}$$
$$Si^{30}(p,\gamma)P^{31}$$

5. Area of Investigation

From initial survey runs, a section of prominent resonance peaks in the unreported range of 1.0 to 2.0 Mev was chosen for investigation. The region from 1.57 to 1.80 Mev was studied in detail.

6. Equipment

The two Mev horizontal Van de Graaff at the U. S. Naval Postgraduate School was used to accelerate the protons. This machine was manufactured by High Voltage Engineering Corporation. The proton source is similar to the radio frequency ion source described by C. D. Moak.[10]

A 25 degree magnetic analyzer was used for determination of the energy of the accelerated protons. The deflection angle was accurately defined by positioning the slit intercepting the analyzed beam a distance of about 2.1 meters from the magnet center. The magnet current was passed through a 0.1 ohm shunt and the potential difference measured by a Leeds and Northrup potentiometer. See Figure 1. No magnetic field determinations were made for the analyzer as proton energies were calibrated to magnet currents by the known resonances in the capture of protons by lithium and

[10] C. D. Moak, et al, Nucleonics, 9, No. 3, 18 (1951)

Figure 1

The potentiometer-galvonometer measuring of magnetic analyzer current.

fluorine as outlined in the section on "Calibration of Proton Energy".

A corona stabilizer was used in conjunction with the accelerator. It is a device whereby a signal voltage taken from a slit intercepting a portion of the analyzed proton beam is amplified and used to control the current flowing between the accelerator terminal and a set of adjustable corona points facing the terminal, thereby regulating the terminal voltage. Because of a Bremsstrahlung radiation of about 300 m.r./hour near the back of the machine, it was convenient to remotely control the corona point settings in order to permit continuous operation over a wide range of energies. This was accomplished by installation of a gear train and selsyn system so that the corona points may be set from the control console. See Figures 2 and 3.

The target and the target chamber were electrically insulated from each other and the remainder of the system. See Figure 4. This permitted the target chamber to be biased with a 300 volt dry cell battery, in series with a 10 megohm resistor to form a Faraday cage. The Faraday cage repels the electrons formed by ionization of residual gas molecules and prevents the escape of electrons from the target which would give a false beam current. The target was then electrically connected to an integrator so that counts could be measured for a specific capacitor charge, i.e. a precise number of protons impinging upon the target. In practice, a relay of the integrator was wired directly to the count switch of a Berkeley Decimal Scaler, Model 2001, so that upon

Figure 2

Gear train and selsyn drive for corona point adjustment

Figure 3

Van de Graaff control console showing remote corona point adjustment selsyn.

Figure 4

Target chamber and scintillation detectors

actuation of the start switch of the integrator, the counter and timer of the scaler would start and also be shut off automatically when the desired voltage on the capacitor was obtained. Also connected to another pair of relay contacts of the integrator was a solenoid-operated beam chopper or shutter.

This shutter was installed to prevent overheating and unwanted bombardment of the target. It was placed between the intercepting slit and the target. See Figure 5. At the times that the integrator allowed the scaler to operate, it also opened the circuit to the coil of the shutter allowing it to drop by gravity so that a hole in the shutter permitted the proton beam to pass to the target. When the integrator stopped the scaler, it simultaneously applied power to the shutter coil causing it to present a solid section of the shutter to the beam. In this method, the beam was restricted from the target without a mechanical opening to the vacuum system and without a magnetic disturbance to the proton beam. (The a.c. shutter coil is not energized while bombarding the target.) Furthermore, a simple on-off switch between the shutter coil and the integrator permits opening of the circuit to bombard the target for focusing. The target current meter was also housed with the integrator.

The target end system shown in Figures 6 and 7 was found most convenient. Between the slits intercepting the analyzed proton beam and the target, a valve and a T fitting were installed. The branch tube thus formed allowed incorporation of an auxiliary oil diffusion and fore pump system.

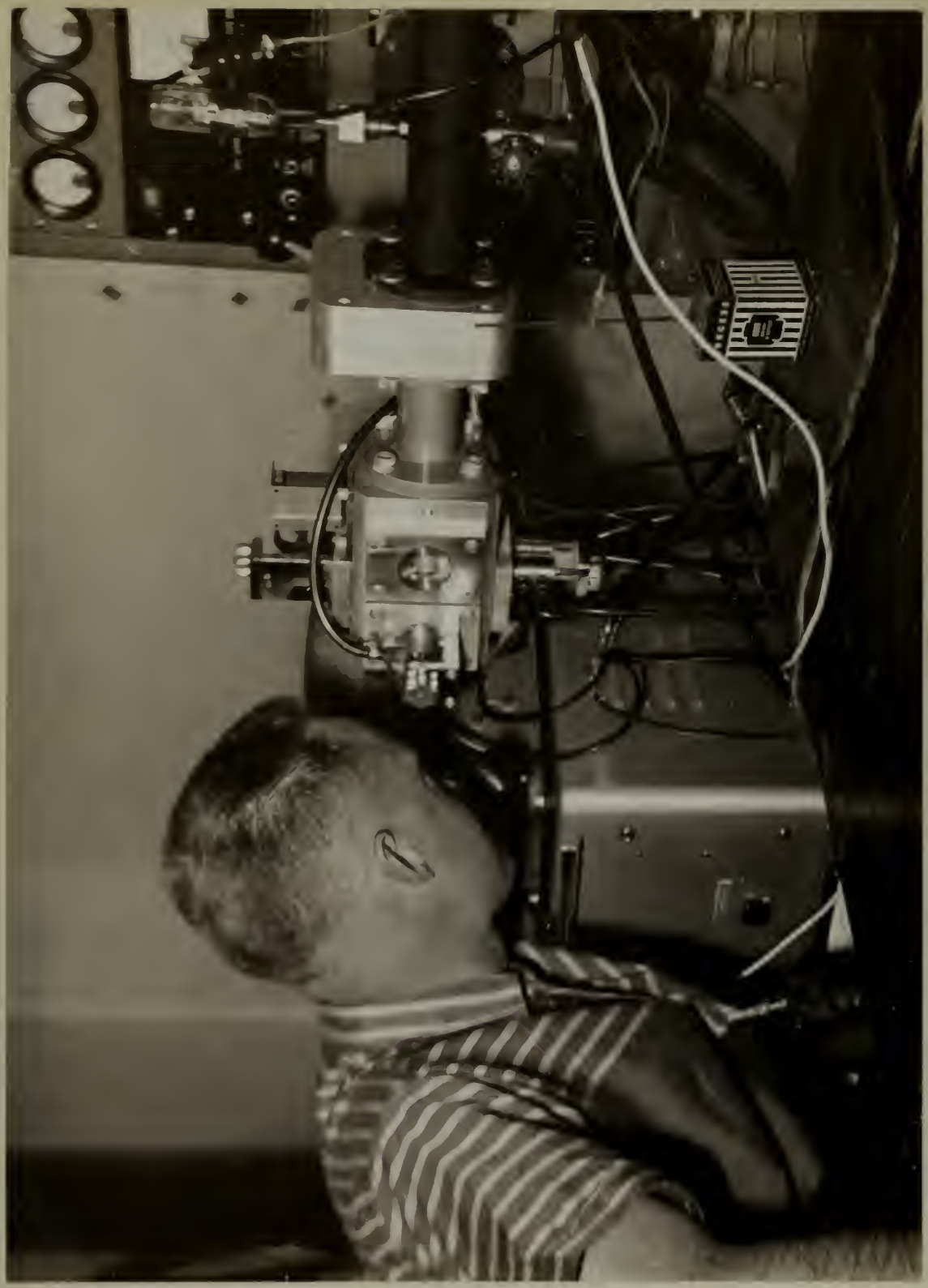


Figure 5

The slit-view box showing addition of solenoid actuated shutter.

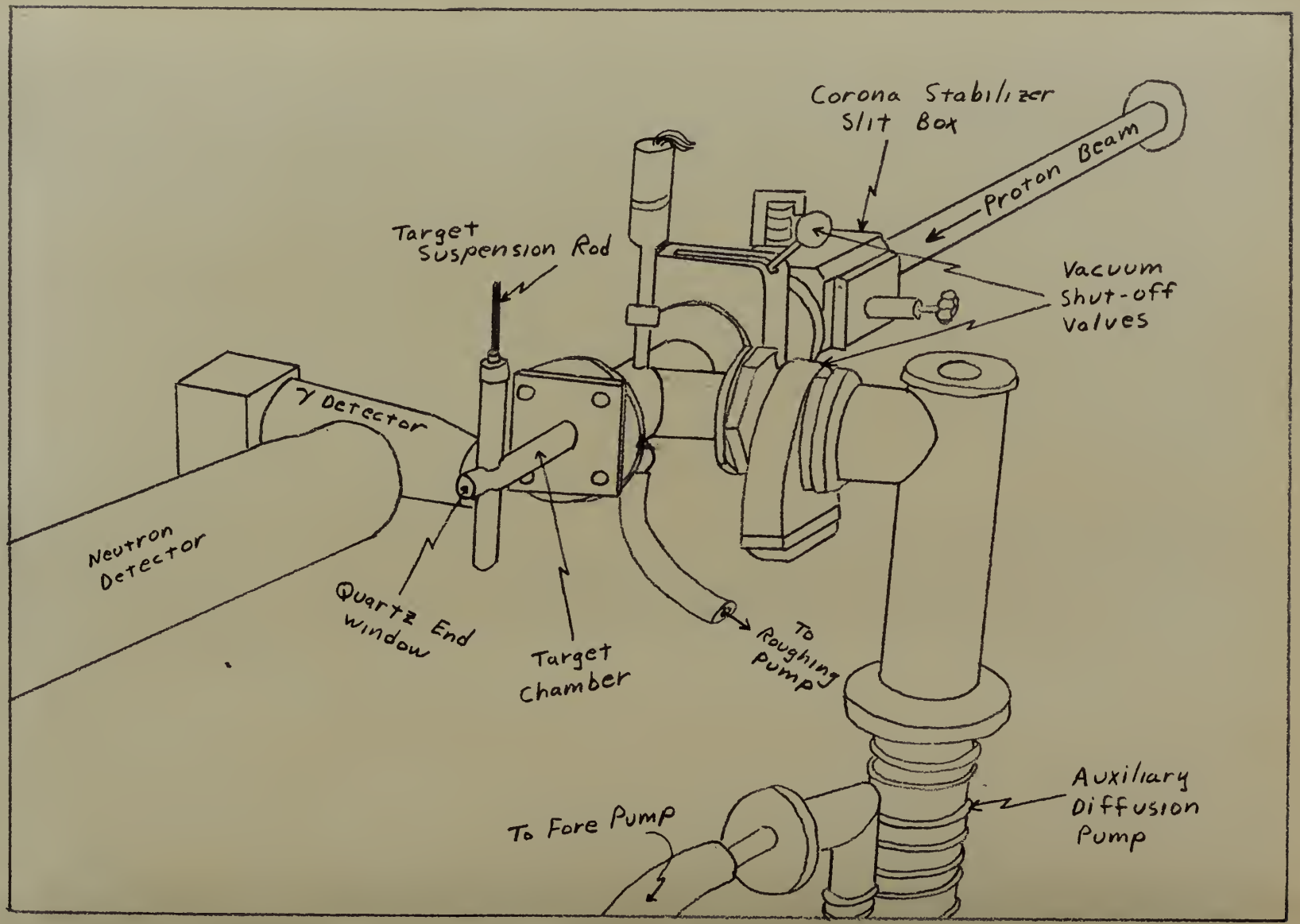

Corona Stabilizer
Slit Box
Proton Beam
Target
Suspension Rod
Vacuum
Shut-off
Valves
γ Detector
Neutron
Detector
Quartz End
window
Target
Chamber
To
Roughing
Pump
Auxiliary
Diffusion
Pump
To Fore Pump

Figure 6

Target End System

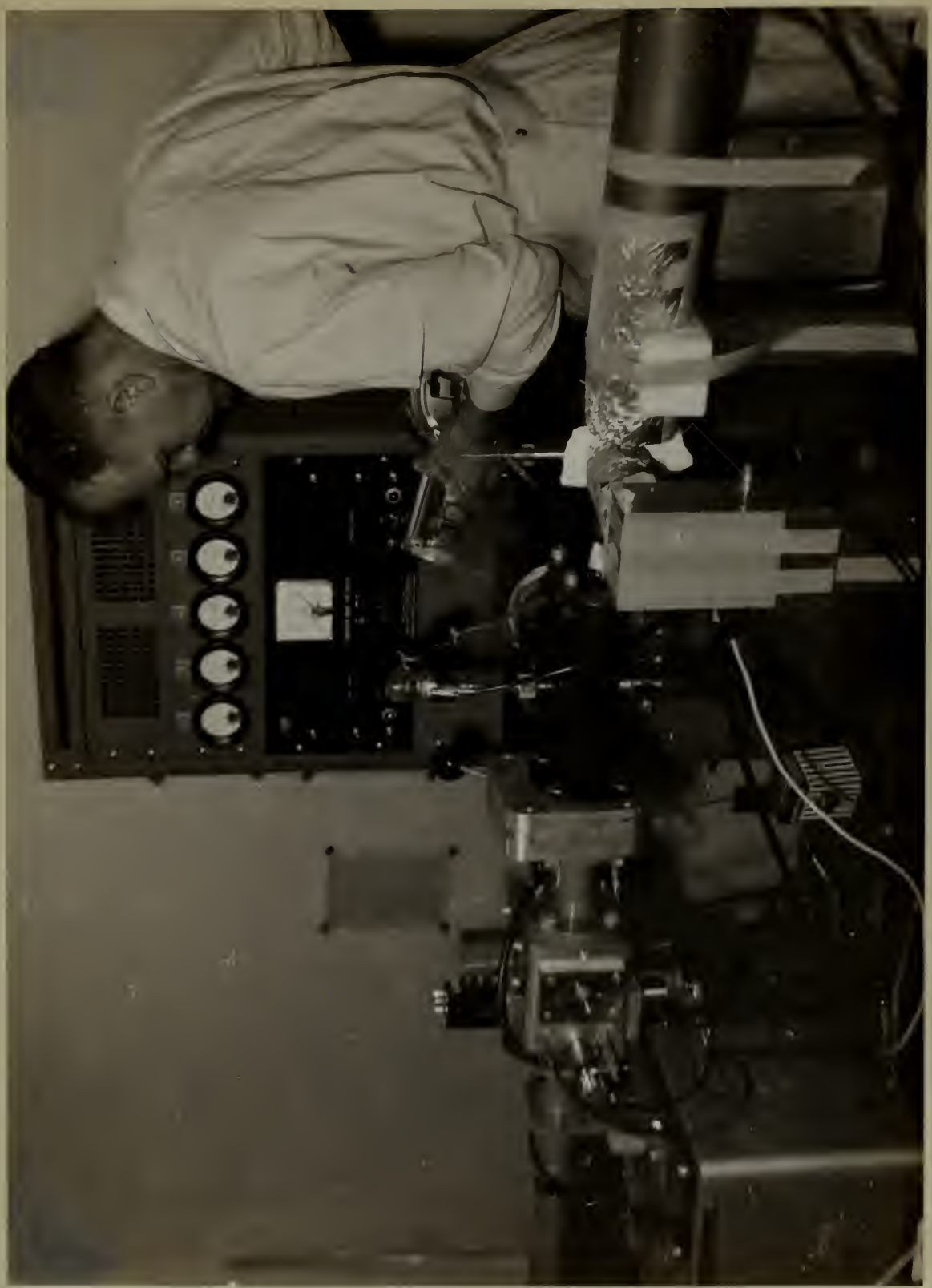


Figure 7

Cooling the auxiliary diffusion pump trap with liquid nitrogen

Another valve allowed isolation of these pumps from the straight through portion of the T fitting. Also incorporated in the T fitting were an outlet for an ion gauge tube and a connection with valve for a roughing pump. By isolating the auxiliary pumps and closing the valve by the slits, only a small volume of vacuum was lost when changing targets. After a new target was installed and the space roughed out, the main valves could be reopened without losing the effectiveness of the diffusion pumps. A change of targets could be made and vacuum restored so that bombardment could continue in less than five minutes. A fused quartz crystal, polished on both surfaces, was used as an end of the target chamber. This was extremely useful, not only for aligning the system, but also as a window for viewing the back of the target to assure proper positioning.

The gamma-ray yield from the reactions was measured by a two-inch diameter thallium-activated, sodium iodide crystal mounted on a Dumont 6292 photomultiplier tube. The photomultiplier signal was amplified and counted as shown on the block diagram, Figure 8. The discriminator was set to prevent counting of all gamma photons of less than about 1.2 Mev energy. Similarly the neutron yield was measured by a one inch diameter europium-activated, lithium iodide crystal mounted on a Dumont 6292 photomultiplier. The amplification and counting block diagram is shown on Figure 8.

7. Target Production

For the thin LiF targets used for calibration of the proton energies, blanks were produced from commercial "Buss

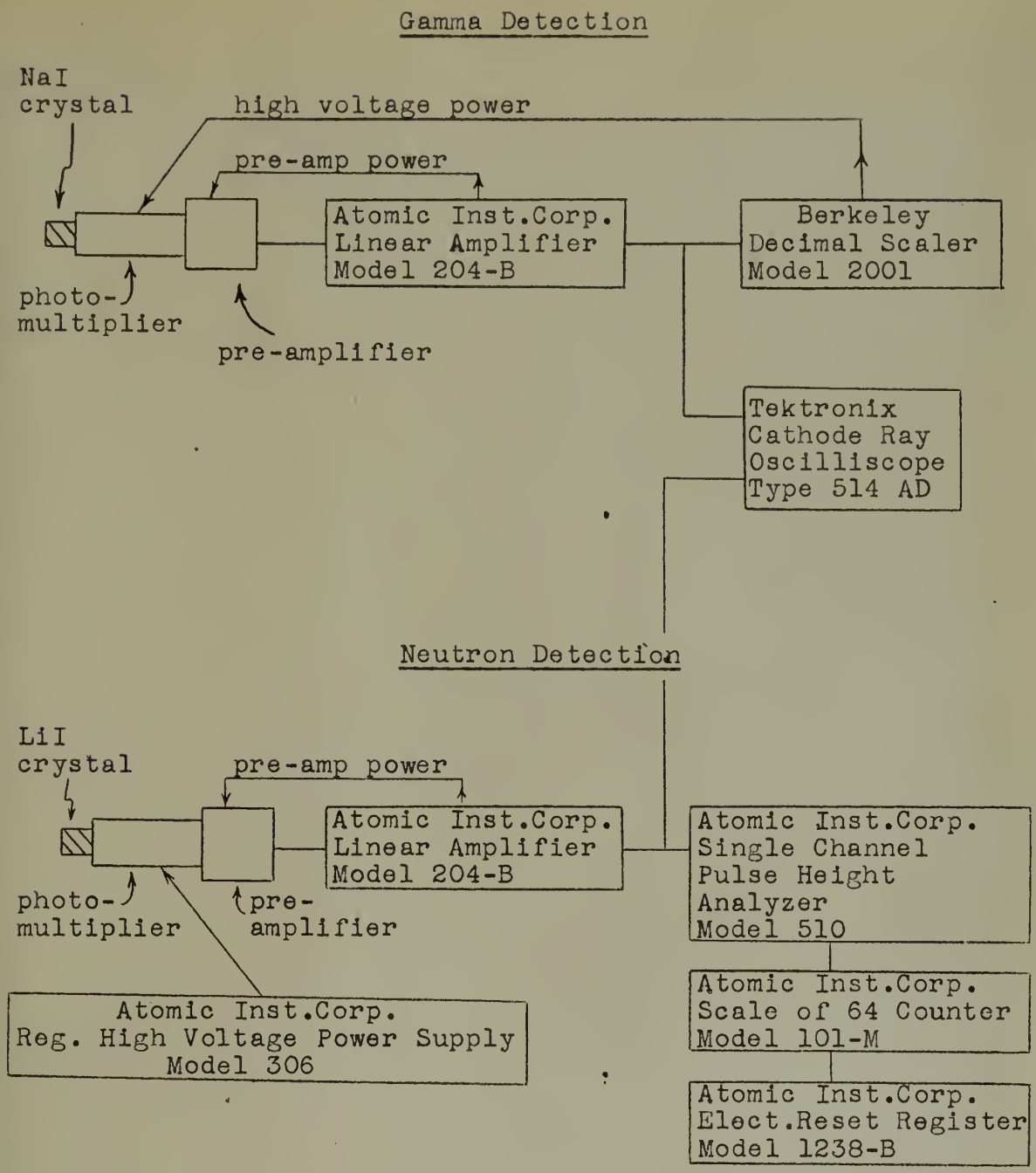
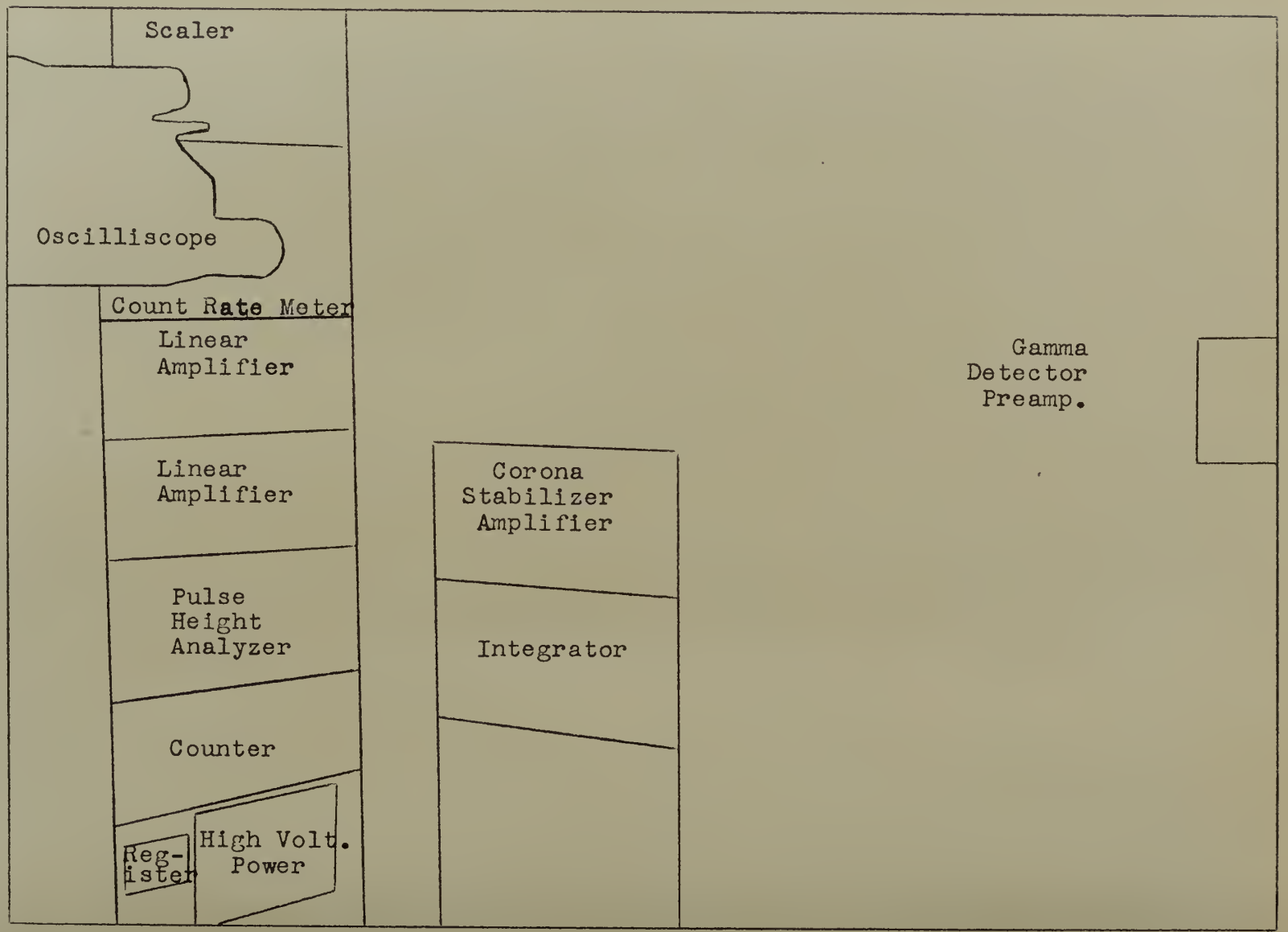


Figure 8

Block diagram of Gamma and Neutron Detection Equipment


Scaler
Oscilliscope
Count Rate Meter
Linear Amplifier
Linear Amplifier
Pulse Height Analyzer
Counter
Reg-ister
High Volt. Power
Corona Stabilizer Amplifier
Integrator
Gamma Detector Preamp.

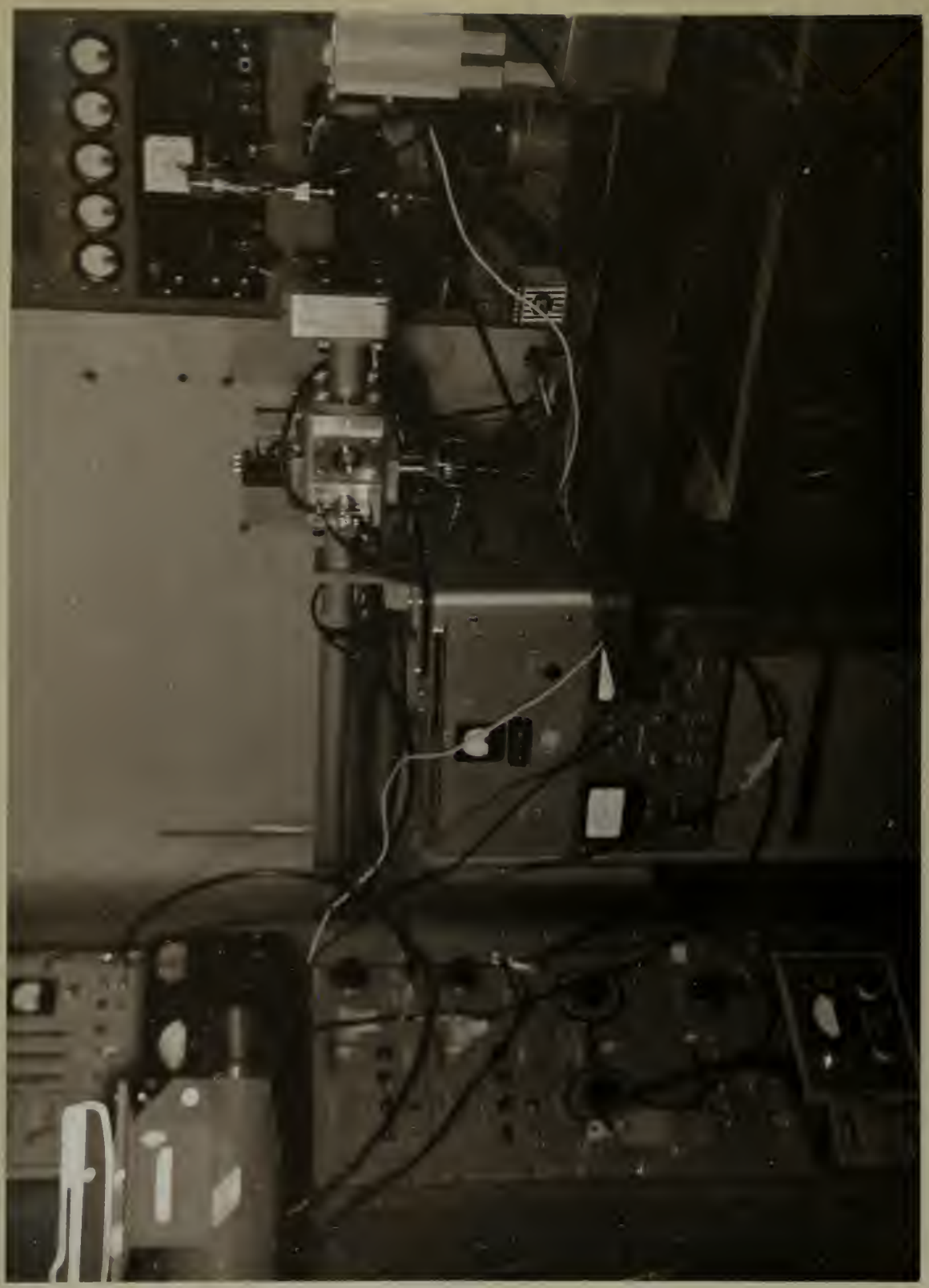


Figure 9

Gamma and neutron detection equipment

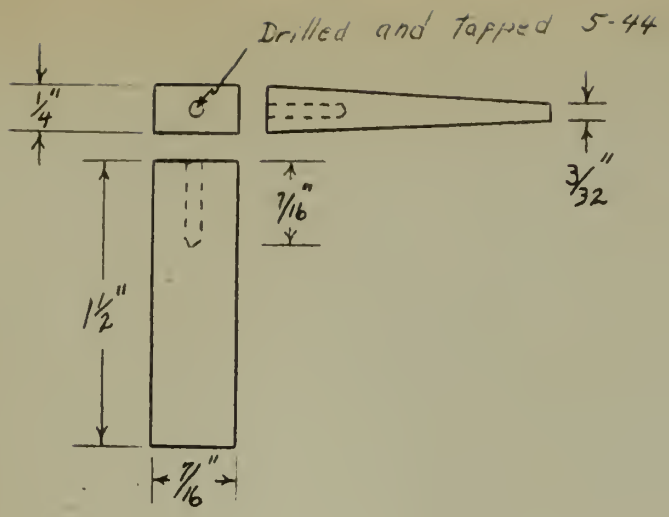


Figure 10

Bar" electrical copper. A blank is shown in Figure 10. The copper produced an increasing background above about one Mev, but this was insufficient even at 1.8 Mev to obscure the strong fluorine resonances or the lithium neutron threshold. Before plating, the target blanks were polished on one side with a water suspension of alpha Al_2O_3 powder to give a clear, mirror-like finish, free of scratches visible to the naked eye. Three targets were vacuum plated with LiF before one was obtained which was sufficiently thin to produce good thin target resonances. The criteria here seems to be: if you are sure you can see a plating of LiF, it is too thick.

For the thin silicon targets, natural SiO was vacuum plated on top of a heavy vacuum plating of silver, using copper target blanks the same as the ones used for the LiF targets. The background radiation for this system proved too great to be acceptable for the small gamma yield obtained. A satisfactory target was then produced by slotting

the bottom of a standard copper blank with a fine jeweler's saw to about 3/32 of an inch depth. A strip of .0005 inch tantalum sheet was inserted in the slit and the copper peaned to securely grip the Ta strip. The tantalum was then stretched over the polished surface of the blank and held at the top with a washer and nut riding on the suspension rod. The tantalum was cleaned with c.p. ethyl alcohol prior to vacuum plating with SiO. Judging from the green color of the plated target it is estimated that the coating obtained was about 15 kev thick.[6]

No completely satisfactory method was devised for production of a thick silicon target. It was desired to use a SiO_2 (fused quartz crystal) for a thick target as proof that the resonances obtained were actually from silicon and not some contaminant. The first of these targets produced was simply the drilling and tapping of the polished face of a copper target blank so that an upper and lower screw head would hold the crystal in place. This crystal shattered after about one-half hour's bombardment. A second crystal was mounted by an upper and lower spring clip to a flat unpolished brass plate of similar dimensions to the copper blanks. This crystal survived nearly two hours of intermittent bombardment. It is not known whether heat or surface charge of the bombarding protons caused failure of the crystals. The nature of the target chamber used did not permit inclusion of a water cooling system to the target backing. Electrical contact to the face of the ends of the

crystal was assured, but these were nearly one-half inch from the point of bombardment.

8. Data Collection

A two man team was used for data collection. One man, stationed at the console, regulated the operation of the Van de Graaff and magnetic analyzer. To minimize the hysteresis effect in the electromagnet, the magnet was slowly cycled twice before each series of readings were made, and further, during any series of readings the magnet current was always increased, and never decreased. This last requirement made adjustment of the magnet current much more painstaking, but the capability of being able to repeat results indicates that this procedure is a good one. The magnet was also operated primarily in the linear region; the magnet is rated at five amperes and the maximum current used was about 1.8 amperes. The potentiometer was checked against a standard cell just prior to each setting of the electromagnet.

The man stationed near the target chamber operated the integrator and detection systems. From here the Van de Graaff focus was controlled, through the intercom to the console, by reference to the target current meter, and similarly the setting of the high potential voltage was co-ordinated by reference to the corona stabilizer balance meter.

9. Calibration of Proton Energy

A thin lithium fluoride target was used for calibration of the proton energies. Complete sweeps of the energy range

from 0.2 Mev to 2.0 Mev were made in sufficiently small steps to allow identification of the prominent fluorine resonances by gamma yield, and the lithium neutron threshold. The fluorine gamma resonances are primarily from the reaction: $F^{19} + p \rightarrow Ne^{20*} \rightarrow O^{16*} + \alpha \rightarrow O^{16} + \gamma$. The proton energies in kev, used for the fluorine, were: 483.1, 669, 831, 873.5, 900, 935.3, 1176, 1290, 1346, 1372, and 1690.[11,12] The proton energy used for the Li^7 (p,n) Be^8 threshold was 1.881 Mev. This provided the information necessary to plot the curve of proton energy versus magnet current shown in Figure 11. It should be noted that the slit was set with a horizontal opening of between 1.0 and 1.5 mm and was not subsequently moved, thus assuring a constant deflection angle by the magnetic analyzer.

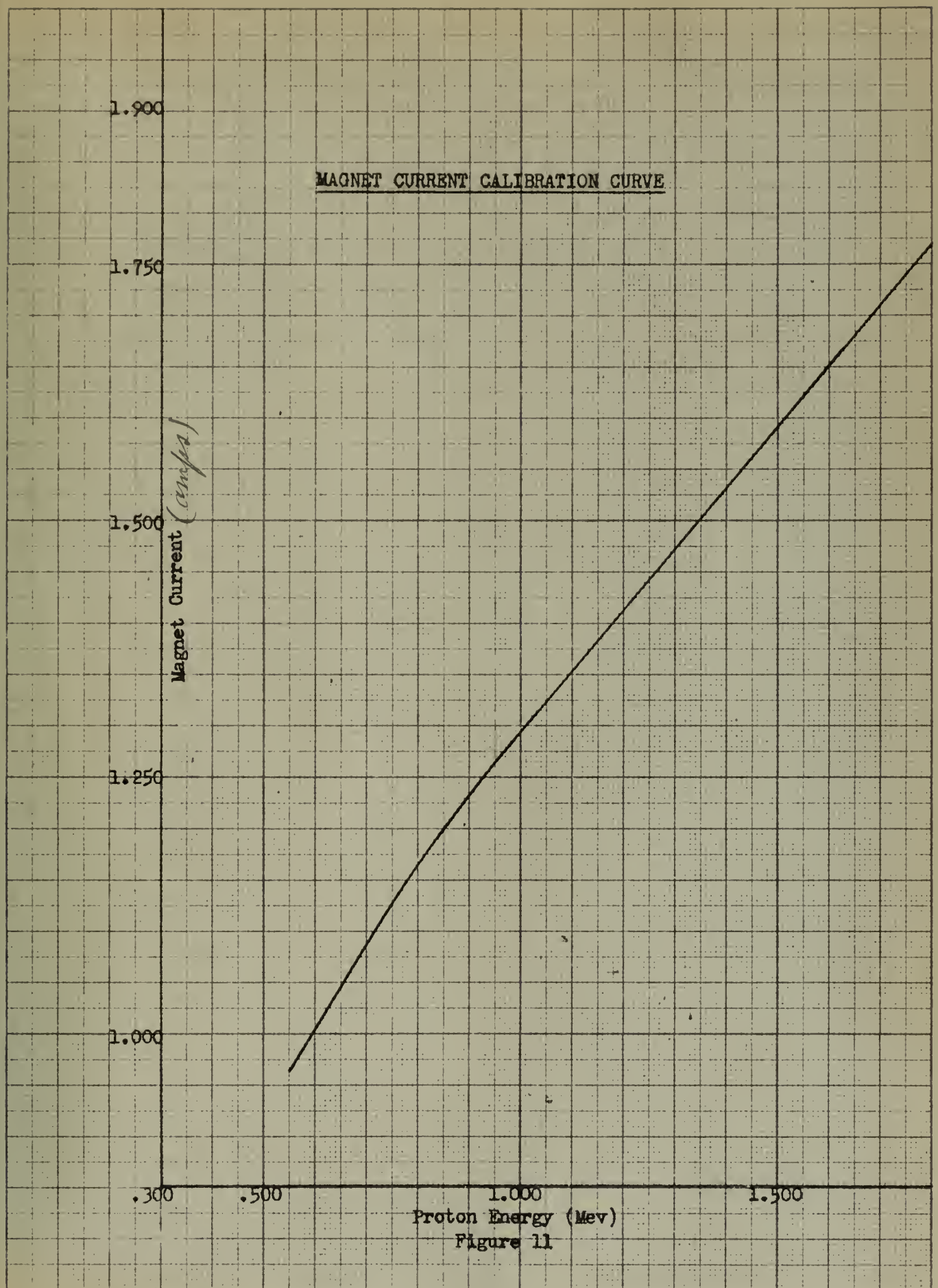
10. Resolution of Equipment

As observed by others using tantalum targets for proton bombardment,[6,11] fluorine resonances appear because of the natural absorption of fluorine by the tantalum. This factor was utilized to ascertain the resolution capabilities of our complete assembly. The leading edge of the 1372 kev fluorine resonance is known to be a smooth curve.[13] Coverage of this peak in magnet current steps of 0.001 amp provided a perfectly

[11] F. Ajzenberg and T. Lauritsen, Rev. Mod. Phys. 27, 153 (1955)

[12] W. F. Hornyak, et al, Rev. Mod. Phys. 22, 356 (1950)

[13] H. B. Willard, et al, Phys. Rev. 85, 849 (1952)


MAGNET CURRENT CALIBRATION CURVE
1.900
1.750
1.500
1.250
1.000
Magnet Current (amps)
.300
.500
1.000
1.500
Proton Energy (Mev)
Figure 11

smooth leading edge indicating accuracy to at least this degree.

In general the stability of the magnetic analyzer current was excellent. The average drift during the interval of bombardment was less than 0.0001 amp, but on rare occasions a drift during bombardment was observed of nearly 0.0005 amp. When such drifts were observed, an average reading was utilized. It should be noted here, that from Figure 11, in the region of 1.7 Mev, an interval of 0.001 amp corresponds to a change of about 1.7 kev.

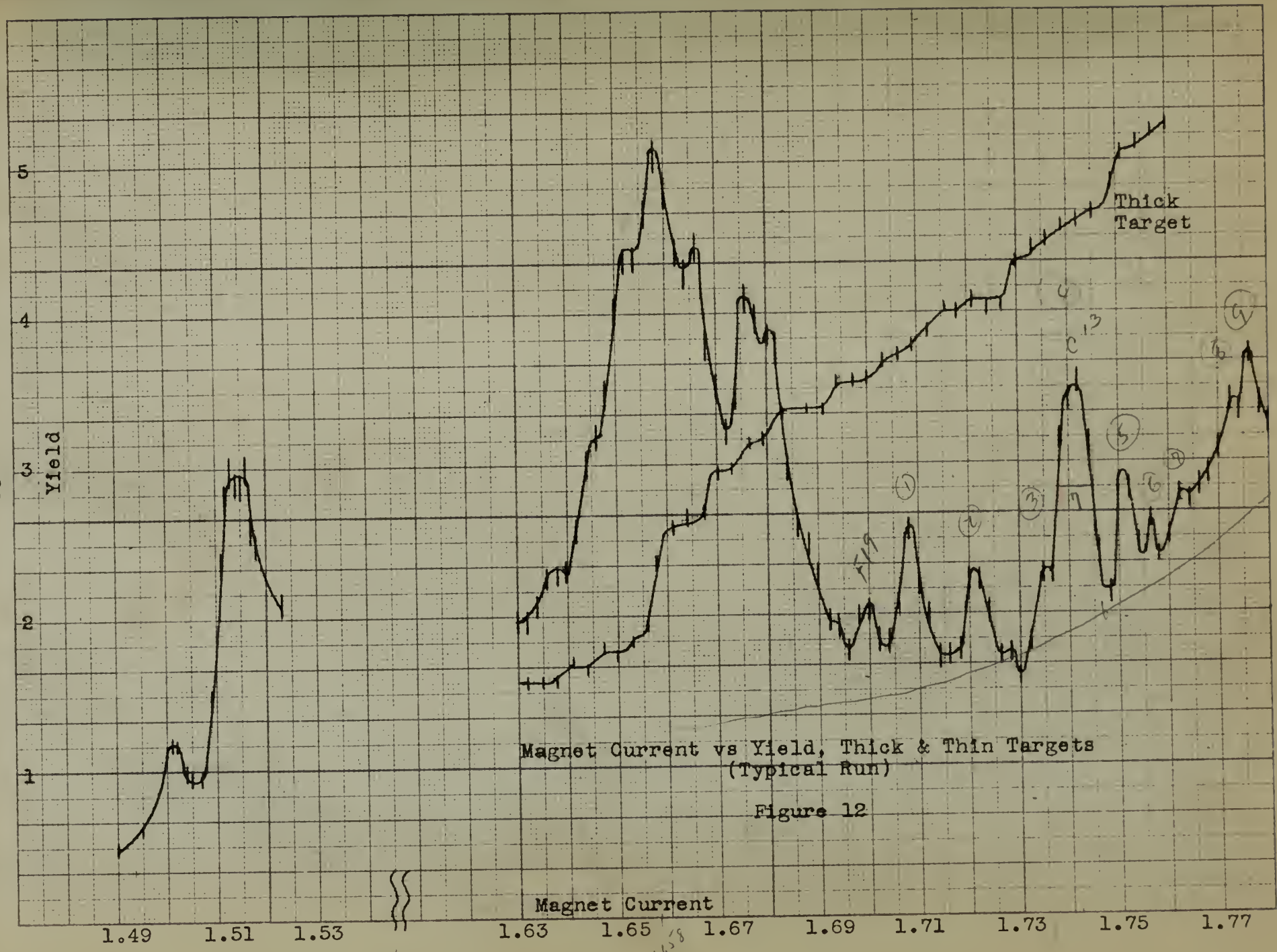
During each coverage of the investigated energy region, the two fluorine resonances at 1372 and 1690 kev were observed. These were used to verify the calibration which for some runs accounted for an offset of nearly 10 kev.

11. Preliminary Survey of SiO

Several preliminary surveys were conducted over the entire range of proton energies from 0.19 to 2.0 Mev. From these surveys, the prominent resonances in the range up to 0.9 Mev previously reported were observed. The surveys also indicated that within the energy capabilities of our equipment, the range from 1.57 to 1.8 Mev held the major interest. Therefore further study was directed toward this latter region.

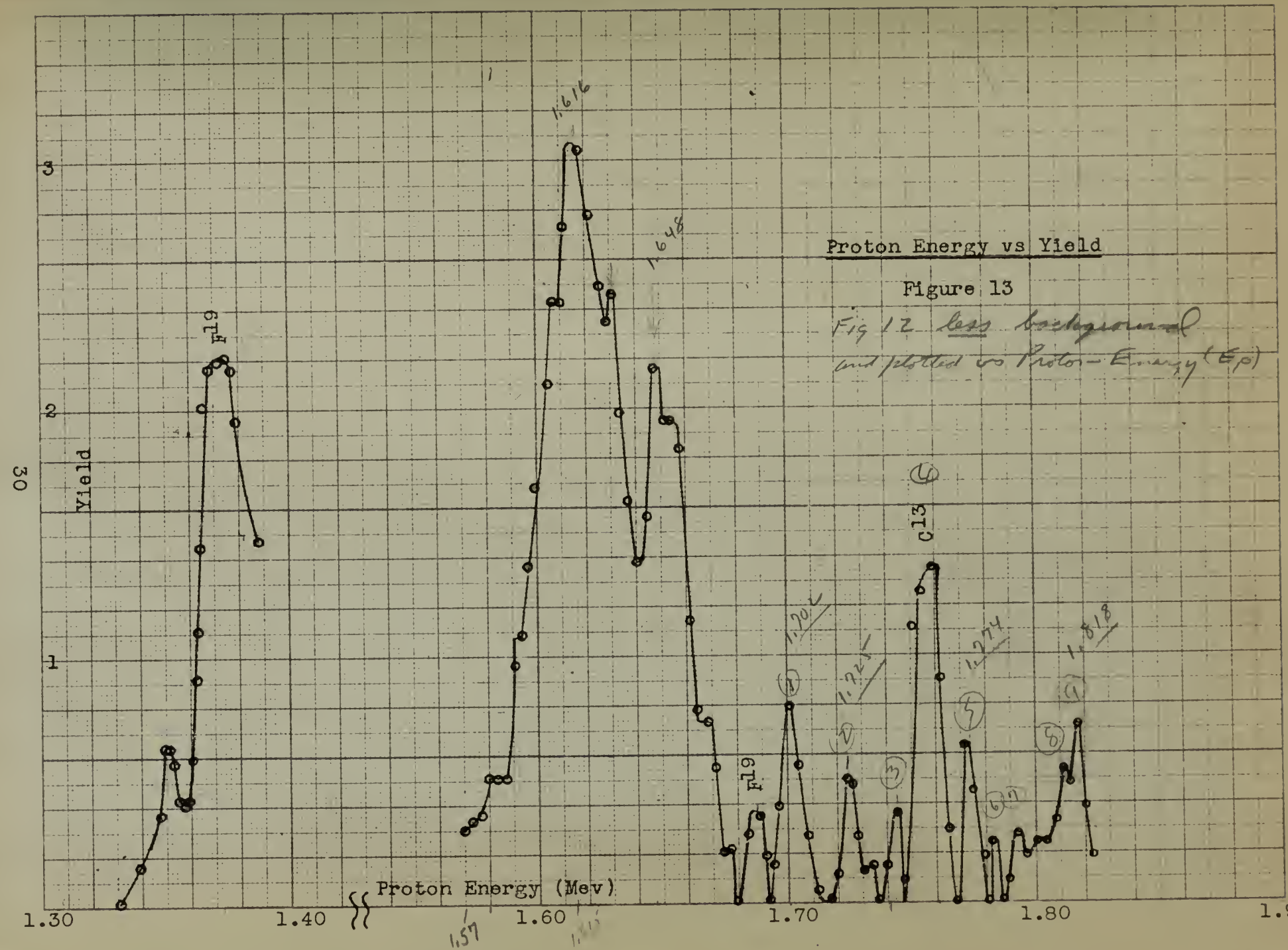
12. Background Radiation

As indicated in Figure 12 a considerable rise in background even with a tantalum target backing was observed over the area of interest. In order to evaluate this rise in



Magnet Current vs Yield, Thick & Thin Targets
(Typical Run)

Figure 12


Proton Energy vs Yield
Figure 13
Fig 12 less background and plotted vs Proton-Energy (Ep)
Yield
Proton Energy (Mev)
1.30
1.40
1.60
1.70
1.80
1.90
1.57
1.616
1.648
F19
F19
C13
1.205
1.274
1.818
1.727
1
2
3
4
5
6
7
8

background radiation special observations were conducted. A target identical to our tantalum-backed SiO target was produced. This plain tantalum target was bombarded over the range of interest to provide background data. This background has been "subtracted off" to produce Figure 13.

13. Silicon verification

To verify that the resonances observed were actually from silicon and not some contaminant, a thick target of SiO_2 was analyzed in the region of interest. The results of these bombardments provided thick target yield curves with steps in the yield at the same energies as resonances were obtained from thin targets. See Figure 12.

14. Results

The more prominent natural silicon resonances of E_p less than one Mev previously reported were observed.

In the range from 1.57 to 1.8 Mev natural silicon resonances were observed as listed in the following table, and shown on Figure 13:

E_p (Mev) ±.002	Relative yield
1.616	420
1.648	298
1.702	111
1.725	72
1.774	86
1.818	99
Possible resonances at:	
1.582	70

1.596	145
1.608	374
1.633	337
1.658	267
1.747	33
1.795	37

No neutron activity was observed at energies up to 2.0 Mev.

The particular silicon isotope causing the listed resonances was not established.

15. Discussion of Errors

A resonance peak was observed at 1.760 Mev with a relative yield of 208. The thick target curve, however, failed to indicate a reasonable slope at this energy. Therefore, it is believed that this resonance is not from silicon but from C^{13}. C^{13} could easily accumulate on the target during bombardment, especially with the oil type auxiliary diffusion pump very near the target chamber.

The accuracy of the proton energies as determined by the manufacturer of the Van de Graaff and magnetic analyzer system was about 0.2 percent at one Mev. Repeated determinations of the resonance points indicates a stability inductive of an error of two kev, corroborating the manufacturer's determination. The final results, however, rest heavily on the accuracy of the 1372 and 1690 kev fluorine resonances which were verified by our calibration runs.

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